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## EUROPEAN PATENT APPLICATION

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54 **Herbicide.**

57 This invention relates to a herbicidal composition comprising fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof or diuron, and to a method of killing weeds by applying to weed seedlings the said combination.

**EP 0 256 785 A2**

HERBICIDE

This invention relates to a new herbicide.

More particularly, it relates to a new herbicidal composition comprising fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof or diuron, and to a method of killing weeds by applying to weed seedlings the said combination.

The fosmidomycin is a known compound, 3-(N-formyl-N-hydroxyamino)propylphosphonic acid as antibacterial agent [Cf. European Journal of Drug Metabolism and Pharmacokinetics Vol. 7, P59 (1982)] and as herbicide [Cf. Japan Kokai No. 106504/1986].

Further, ametryn and diuron are also known herbicides, 2-methylmercapto-4-ethylamino-6-isopropylamino-s-triazine and 3-(3,4-Dichlorophenyl)-1,1-dimethylurea, respectively [Cf. The Merck Index tenth edition items 392 and 3400 (1983)].

The fundamental physiological action of fosmidomycin resides in the inhibition of production of chlorophyll. Therefore, plants emerging after treatment with fosmidomycin are ready to undergo chlorosis. When the treating concentration is such that this chlorosis lasts as long as more than 2 to 3 weeks, arrest of growth occurs as the plant is prevented from nursing itself by photosyntheses, leading to decay. However, as the treating concentration is decreased, the degree and duration of chlorosis are lessened and the plant will not die but show a recovery of growth so that the object of killing cannot be accomplished. While a large variety of herbicides have been developed and put to use for controlling the weeds detrimental to crop plants and the environment, each of these herbicides has its own drawback or shortcoming and none has ever proved fully satisfactory in weed killing effect.

Thus, what are mainly desired in herbicides are:

(1) The maximum possible coverage of weed varieties that can be controlled (a broad herbicidal spectrum)

(2) Manifestation of sufficient control effect within the shortest possible time after application (a reduced number of days required for complete weed control)

(3) No variation in effect according to the volume and height of weeds or an improved stability of control effect (stabilized weed control effect).

(4) Reduced amounts of active substances required for control (reduced herbicide consumption)

The present inventors discovered that application of a composition containing fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and diuron to a plant resulted in a surprisingly great synergistic herbicidal effect on the plant. This finding was followed by a further investigation, which culminated in the present invention.

The salt of fosmidomycin may include an agronomically acceptable salt thereof such as a base salt (e.g. sodium salt, potassium salt, calcium salt, etc.) and the like.

The salt of ametryn may include an agronomically acceptable salt thereof such as an acid addition (e.g. hydrochloride, sulfate, phosphate, etc.) and the like.

The herbicidal composition according to the present invention displays remarkable efficacy as a postemergence herbicide, and is preferably applied to the whole stalks and foliage of weeds that have emerged.

Moreover, the herbicidal composition of the present invention provides effective control, irrespective of weed variety, e.g. broad-leaved weeds and grasses.

The application rate for the active ingredients in the herbicidal composition of the invention varies according to the combination used and kinds of weeds to be controlled. Generally however, the optimum rate of application is selected from the range of 5 to 1000 grams/10 ares and preferably from the range of 100 to 500 grams/10 ares.

The ratio of the fosmidomycin or a salt thereof to the ametryn or a salt thereof or diuron in the herbicidal composition of the invention is dependent on the kinds of respective compounds and the kinds of weeds to be controlled.

Generally, the ratio is selected from the range of 10:1 to 1:10, preferably 6:1 to 1:6.

To apply the herbicidal composition of the invention, it can be mixed with a carrier suited to the intended usage and applied in such varied forms as dusts, granular preparations, wettable powders, liquid preparations, emulsifiable concentrates, flowable emulsion concentrates and so on. The carrier mentioned just above may be a solid or a liquid carrier or a combination thereof. As examples of said carrier, there may be mentioned finely divided minerals such as kaolinite, bentonite, pyrophyllite, talc, diatomaceous earth, silica gel, calcium carbonate, etc., finely divided vegetable materials such as starch, gum arabic, etc.,

organic solvents such as alcohols, ketones, kerosine, benzene, toluene, xylene, cyclohexane, methylnaphthalene, dioxane, dimethylformamide, dimethyl sulfoxide, corn oil, o-dichlorobenzene, isophorone, water and so on. Further, agronomically acceptable adjuvants and auxiliaries such as wetting agents, dispersing agents, adhesives, extenders, etc. can be incorporated, if necessary, in appropriate proportions.

Each of these preparations is not only useful as such but may be used in combination with bactericides, fungicides, nematocides, insecticides, plant growth regulators, fertilizers, other herbicides and so on.

The following reference and working examples are intended to illustrate the invention in further detail.

#### 10 Reference Example

Seeds of the following species of plants were sown in soil-filled pots (30 cm long × 10 cm wide × 10 cm deep) and grown in a glasshouse. After 2 weeks, seedlings which had grown to a height of 5 to 20 cm were treated with test compound. The test compound was dissolved in water and the solution was adjusted to a application concentration of 500 ppm or 5000 ppm and applied to the whole surface of the plant at the rate of 100 liters water/10 ares. Control pots were treated with water only. After 10, 20 and 30 days following application, the plants were observed for growth and the control effect of the test compound was evaluated.

The test plants were as follows.

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Test plant

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No. Common name

Botanical name

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1 Rice

Oryza sativa L.

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	2	Maize	Zea mays L.
5	3	Barnyard millet	Panicum Crus-galli L.
	4	Crabgrass	Digitaria adscendes Henr.
10			
	5	Soybean	Glycine Max MERR.
15	6	Convolvulus	Calystegia japonica choisy
20	7	Barnyard grass	Amaranthus Blitum L

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The test compound is as follows.

30	Compound No.	Chemical name
	1	3-(N-Formyl-N-hydroxyamino)propyl-
35		phosphonic acid monosodium salt

40 The test results are set forth below in the table. Control effects were scored on a rating scale of 0 for no effect through 100 for complete control (complete kill).

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Com- pound No. (concentra- tion)	Time after appli- cation in days	Plant No.	Herbicidal activity (%)						
			1	2	3	4	5	6	7
1 (500ppm)	10		(22)	(27)	(34)	(40)	(46)	(25)	(28)
	20		(56)	(44)	(61)	(67)	(60)	(40)	(43)
	30		38	31	55	52	42	30	27
1 (5000ppm)	10		(51)	(65)	(72)	(78)	(59)	(52)	(69)
	20		100	100	100	100	100	100	100
	30		100	100	100	100	100	100	100

The open circle around each figure indicates the occurrence of chlorosis.

### 30 Example 1

Using the same test plant species and the same test procedure as described in Reference Example, the control effects of the following compounds were tested and evaluated.

The observation of plants for growth status was made on 10 days after application.

35 The test compounds are as follows.

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Compound No.	Chemical name (concentration)
1	2-Methylmercapto-4-ethylamino-6-isopropylamino -s-triazine 1*) (500 ppm)
2	2-Methylmercapto-4-ethylamino-6-isopropylamino -s-triazine 1*) (500 ppm) + 3-(N-Formyl-N-hydroxyamino)propylphosphonic acid monosodium salt (500 ppm)
3	3-(N-Formyl-N-hydroxyamino)propylphosphonic acid monosodium salt (500 ppm)
Note)	As test compounds 1*), the corresponding commercial product Gesapax (25% emulsifiable concentrate, Ciba-Geigy) was used.

The test results are as shown below in the table.

Com- pound No.	Plant No.	Hibicidal activity (%)						
		1	2	3	4	5	6	7
1 (Control)		78	82	100	100	100	80	78
2		100	100	100	100	100	100	100
3 (Control)		(22)	(27)	(34)	(40)	(46)	(25)	(28)

The open circle around each figure indicates the occurrence of chlorosis.

Example 2

The field on which the following weeds were grown, were plotted out in a ratio of 4m<sup>2</sup>/lot.

The following test compound was dissolved in water at the specified application rate and was sprayed onto the whole surface of the weeds at the volume rate of 100 liters water/10ares.

Control lots were treated with water only.

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Test weed code	Test weeds common name	Botanical name	height (cm)
a	Fleabane, Annul	Erigeron annuus L.	60
b	Ohinutade	Polygonum Blumei Meisn	60

5	Test weed code	Test weeds common name	Botanical name	height (cm)
	c	Clover, Red	Trifolium protense L.	40
10	d	Mugwort	Artemisia vulgaris L. var. indica Maxim	40
15	e	Dandelion	Taraxacum Officinale Weber	10-20
20	f	Brome, Rescue	Bromus unioloides Humb. Bonpl. et Kunth	60
25				

30	Test compound No.	Chemical name
35	A	3-(N-Formyl-N-hydroxyamino)- propylphosphonic acid monosodium salt
40	B	2-Methylmercapto-4-ethylamino-6- isopropylamino-s-triazine 1**)
45	C	3-(3,4-Dichlorophenyl)-1,1- dimethylurea 2**)

50      Note) As test compounds 1\*\*) and 2\*\*), the  
corresponding commercial products, Gesapax (25%  
emulsifiable concentrate, Ciba-Geigy) and Karmex  
55      D(78.5% wettable powder, du Pont) were used,  
respectively.



Two weeks after the treatment, herbicidal effect of the test compounds on the weeds were observed and scored on a rating scale of 0 for no effect through - 100 for complete kill.

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Test compound and application rate (g/10a)		Test weed and herbicidal activity (%)					
B	A	a	b	c	d	e	f
0	25	5	5	5	10	10	0
0	50	10	10	10	15	10	10
0	100	20	20	20	30	15	10
62.5	0	0	0	0	0	0	0
125	0	20	20	15	20	10	15
250	0	40	50	60	60	60	55
62.5	25	60	60	50	60	70	60
125	25	70	65	70	80	70	70
250	25	85	80	85	80	85	80
62.5	50	70	70	75	80	70	70
125	50	80	80	90	90	80	80
250	50	90	>90	90	>90	90	90
62.5	100	80	90	80	>90	80	80
125	100	90	>90	>90	>90	90	90
250	100	>90	>90	>90	>90	>90	>90

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5	Test compound and application rate (g/10a)		Test weed and herbicidal activity (%)					
	C	A	a	b	c	d	e	f
10	0	25	5	5	5	10	10	0
	0	50	10	10	10	25	10	10
	0	100	20	20	20	30	15	10
15	250	0	5	5	10	5	10	5
	500	0	15	10	20	15	20	15
	750	0	40	30	40	30	40	30
20	250	25	40	50	30	50	50	40
	500	25	60	60	40	50	55	60
	750	25	85	80	75	85	85	85
25	250	50	60	70	50	70	60	60
	500	50	80	90	80	90	90	85
	750	50	90	90	90	>90	90	90
30	250	100	85	90	80	90	80	70
	500	100	90	>90	>90	>90	>90	>90
	750	100	>90	>90	>90	>90	>90	>90
35	250	100	85	90	80	90	80	70
	500	100	90	>90	>90	>90	>90	>90
	750	100	>90	>90	>90	>90	>90	>90
40	250	100	85	90	80	90	80	70
	500	100	90	>90	>90	>90	>90	>90
	750	100	>90	>90	>90	>90	>90	>90

### 45 Example 3

Seeds of goosefoot were sown in soiled polyethylene pots (20 cm long × 10 cm wide × 10 cm deep) and grown in a glasshouse. Numbers of seedlings (Grown height : 30 cm) were adjusted to 10/pot. Each test compound was dissolved in water at the specified application rate and applied onto the leaves and stocks of Goosefoot. The treated goosefoot were grown in a glass house at 25-30°C for 2 weeks. The weight of the remaining and living portion (above ground) of goose foot was determined and from the obtained data, ratio (%) of the weight of treatment group to that of non-treatment group was calculated. The results are shown in the following table (a).

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(a) Ratio (%) of the remaining weight of the treated plant to that of the non-treated plant

		application rate (g/10a) of the test compound A			
		0	25	50	100
application rate (g/10a) of the test compound B	0	100	95	85	85
	62.5	100	45	25	10
	125	80	20	10	<10
	250	50	10	10	<10

Note: Test compound A: 3-(N-formyl-N-hydroxyamino)-propylphosphonic acid monosodium salt

Test compound B: 2-methylmercapto-4-ethylamino-

6-isopropylamino-s-triazine  
[Gesapax (25% emulsifiable concentrate, Ciba-Geigy)]

The above data were analyzed according to Colby's equation (cf. weeds, Vol, 15, P. 20-22).

Expected value (%) of the remaining weight of the  
plant treated with the combination

Observed value (%) of the  
remaining weight of the  
plant treated with  
compound A only

X

Observed value (%) of  
the remaining weight  
of the plant treated  
with compound B only

=

100

In case that the expected value (%) is larger than the observed value (%), the combination can be judged to have synergistic effect.

The Colby's expected values (%) obtained by calculating using the above data are shown in the following table (b).

(b) Colby's expected value

		application rate (g/10a) of the test compound A		
		25	50	100
application rate (g/10a) of the test compound B	62.5	95	85	85
	125	76	68	68
	250	47.5	42.5	42.5

As clear from the data listed in the above table (a) and (b), all combinations of the test compound A and B can be judged to have synergistic herbicidal activity.

## Example 4

The following data were obtained in a similar manner to those of Example 3, excepting the test compound C was used in place of the compound B.

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(a) Ratio (%) of the remaining weight of the treated plant to that of the non-treated plant

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		application rate (g/10a) of the test compound A			
		0	25	50	100
application rate (g/10a) of the test compound C	0	100	95	85	85
	250	95	60	40	20
	500	85	45	10	<10
	750	60	20	10	<10

Note: Test compound A is the same compound as defined in Example 3.

Test compound C : 3-(3,4-dichlorophenyl)-1,1-dimethylurea  
[Karmex D (78.5% wettable powder, du Pont)].

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## (b) Colby's expected value (%)

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		application rate (g/10a) of the test compound A		
		25	50	100
application rate (g/10a) of the test compound C	250	90	81	81
	500	81	72	72
	750	57	51	51

As clear from the data listed in the above table (a) and (b), all combinations of the test compound A and C can be judged to have synergistic herbicidal activity.

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**Claims**

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1. A herbicidal composition comprising fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and diuron, and agronomically acceptable carrier(s).
2. The herbicidal composition of claim 1, which comprises fosmidomycin or salt thereof in combination with ametryn or salt thereof, and agronomically acceptable carrier(s).
3. The herbicidal composition of claim 1, which comprises fosmidomycin or salt thereof in combination with diuron, and agronomically acceptable carrier(s).
4. A method of killing weeds which comprises applying to weed seedlings fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and diuron.

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(54) **Herbicide.**

(57) This invention relates to a herbicidal composition comprising fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof or diuron, and to a method of killing weeds by applying to weed seedlings the said combination.

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# EUROPEAN SEARCH REPORT

Application Number

EP 87 30 6960

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	US-A-4 206 156 (T. KAMIYA et al.) * Claims * ---	1-4	A 01 N 57/20 // (A 10 N 57/20
P,A	CHEMICAL ABSTRACTS, vol. 105, no. 19, 10th November 1986, page 261, no. 166897j, Columbus, Ohio, US; & JP-A-61 106 504 (TEIJIN LTD) 24-05-1986 (Cat. D,A) * Abstract * ---	1-4	A 10 N 47:30 A 10 N 43:70 )
A	C.R. WORTHING et al.: "Pesticide Manual", 7th edition, pages 226,250, 1983, British Crop Protection Council, Croydon, GB; * Page 11, entry 250; page 226, entry 5400 * -----	1-4	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			A 01 N
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 12-09-1988	Examiner FLETCHER A.S.
<b>CATEGORY OF CITED DOCUMENTS</b>			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document	

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A	US-A-4 206 156 (T. KAMIYA et al.) * Claims * ---	1-4	A 01 N 57/20 // (A 10 N 57/20
P,A	CHEMICAL ABSTRACTS, vol. 105, no. 19, 10th November 1986, page 261, no. 166897j, Columbus, Ohio, US; & JP-A-61 106 504 (TEIJIN LTD) 24-05-1986 (Cat. D,A) * Abstract * ---	1-4	A 10 N 47:30 A 10 N 43:70 )
A	C.R. WORTHING et al.: "Pesticide Manual", 7th edition, pages 226,250, 1983, British Crop Protection Council, Croydon, GB; * Page 11, entry 250; page 226, entry 5400 * -----	1-4	
			TECHNICAL FIELDS SEARCHED (Int. Cl. 4)
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Further, ametryn and diuron are also known herbicides, 2-methylmercapto-4-ethylamino-6-isopropylamino-s-triazine and 3-(3,4-Dichlorophenyl)-1,1-dimethylurea, respectively [Cf. The Merck Index  
10 tenth edition items 392 and 3400 (1983)].

The fundamental physiological action of fosmidomycin resides in the inhibition of production of chlorophyll. Therefore, plants emerging after treatment with fosmidomycin are ready to undergo chlorosis. When the treating concentration is such that this chlorosis lasts as long as more than 2 to 3 weeks, arrest of growth occurs as the plant is prevented from nursing itself by photosyntheses, leading to decay. However,  
15 as the treating concentration is decreased, the degree and duration of chlorosis are lessened and the plant will not die but show a recovery of growth so that the object of killing cannot be accomplished. While a large variety of herbicides have been developed and put to use for controlling the weeds detrimental to crop plants and the environment, each of these herbicides has its own drawback or shortcoming and none has ever proved fully satisfactory in weed killing effect.

20 Thus, what are mainly desired in herbicides are:

(1) The maximum possible coverage of weed varieties that can be controlled (a broad herbicidal spectrum)

(2) Manifestation of sufficient control effect within the shortest possible time after application (a reduced number of days required for complete weed control)

25 (3) No variation in effect according to the volume and height of weeds or an improved stability of control effect (stabilized weed control effect).

(4) Reduced amounts of active substances required for control (reduced herbicide consumption)

The present inventors discovered that application of a composition containing fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and  
30 diuron to a plant resulted in a surprisingly great synergistic herbicidal effect on the plant. This finding was followed by a further investigation, which culminated in the present invention.

The salt of fosmidomycin may include an agronomically acceptable salt thereof such as a base salt (e.g. sodium salt, potassium salt, calcium salt, etc.) and the like.

35 The salt of ametryn may include an agronomically acceptable salt thereof such as an acid addition (e.g. hydrochloride, sulfate, phosphate, etc.) and the like.

The herbicidal composition according to the present invention displays remarkable efficacy as a postemergence herbicide, and is preferably applied to the whole stalks and foliage of weeds that have emerged.

40 Moreover, the herbicidal composition of the present invention provides effective control, irrespective of weed variety, e.g. broad-leaved weeds and grasses.

The application rate for the active ingredients in the herbicidal composition of the invention varies according to the combination used and kinds of weeds to be controlled. Generally however, the optimum rate of application is selected from the range of 5 to 1000 grams/10 ares and preferably from the range of 100 to 500 grams/10 ares.

45 The ratio of the fosmidomycin or a salt thereof to the ametryn or a salt thereof or diuron in the herbicidal composition of the invention is dependent on the kinds of respective compounds and the kinds of weeds to be controlled.

Generally, the ratio is selected from the range of 10:1 to 1:10, preferably 6:1 to 1:6.

50 To apply the herbicidal composition of the invention, it can be mixed with a carrier suited to the intended usage and applied in such varied forms as dusts, granular preparations, wettable powders, liquid preparations, emulsifiable concentrates, flowable emulsion concentrates and so on. The carrier mentioned just above may be a solid or a liquid carrier or a combination thereof. As examples of said carrier, there may be mentioned finely divided minerals such as kaolinite, bentonite, pyrophyllite, talc, diatomaceous earth, silica gel, calcium carbonate, etc., finely divided vegetable materials such as starch, gum arabic, etc.,

organic solvents such as alcohols, ketones, kerosine, benzene, toluene, xylene, cyclohexane, methylnaphthalene, dioxane, dimethylformamide, dimethyl sulfoxide, corn oil, o-dichlorobenzene, isophorone, water and so on. Further, agronomically acceptable adjuvants and auxiliaries such as wetting agents, dispersing agents, adhesives, extenders, etc. can be incorporated, if necessary, in appropriate proportions.

- 5 Each of these preparations is not only useful as such but may be used in combination with bactericides, fungicides, nematocides, insecticides, plant growth regulators, fertilizers, other herbicides and so on.

The following reference and working examples are intended to illustrate the invention in further detail.

#### 10 Reference Example

- Seeds of the following species of plants were sown in soil-filled pots (30 cm long × 10 cm wide × 10 cm deep) and grown in a glasshouse. After 2 weeks, seedlings which had grown to a height of 5 to 20 cm were treated with test compound. The test compound was dissolved in water and the solution was adjusted to a application concentration of 500 ppm or 5000 ppm and applied to the whole surface of the plant at the rate of 100 liters water/10 ares. Control pots were treated with water only. After 10, 20 and 30 days following application, the plants were observed for growth and the control effect of the test compound was evaluated.

The test plants were as follows.

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Test plant		
No.	Common name	Botanical name
1	Rice	Oryza sativa L.

2	Maize	Zea mays L.
5	3 Barnyard millet	Panicum Crus-galli L.
	4 Crabgrass	Digitaria adscendes Henr.
10		
	5 Soybean	Glycine Max MERR.
15	6 Convolvulus	Calystegia japonica choisy
	7 Barnyard grass	Amaranthus Blitum L
20		

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The test compound is as follows.

30	Compound No.	Chemical name
	1	3-(N-Formyl-N-hydroxyamino)propyl-
35		phosphonic acid monosodium salt

40 The test results are set forth below in the table. Control effects were scored on a rating scale of 0 for no effect through 100 for complete control (complete kill).

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Com- pound No. (concentra- tion)	Time after appli- cation in days	Plant No. Herbicidal activity (%)						
		1	2	3	4	5	6	7
1 (500ppm)	10	(22)	(27)	(34)	(40)	(46)	(25)	(28)
	20	(56)	(44)	(61)	(67)	(60)	(40)	(43)
	30	38	31	55	52	42	30	27
1 (5000ppm)	10	(51)	(65)	(72)	(78)	(59)	(52)	(69)
	20	100	100	100	100	100	100	100
	30	100	100	100	100	100	100	100

The open circle around each figure indicates the occurrence of chlorosis.

### 30 Example 1

Using the same test plant species and the same test procedure as described in Reference Example, the control effects of the following compounds were tested and evaluated.

The observation of plants for growth status was made on 10 days after application.

35 The test compounds are as follows.

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Compound No.	Chemical name (concentration)
1	2-Methylmercapto-4-ethylamino-6-isopropylamino -s-triazine (500 ppm) 1*)
2	2-Methylmercapto-4-ethylamino-6-isopropylamino -s-triazine (500 ppm) 1*) + 3-(N-Formyl-N-hydroxyamino) propylphosphonic acid monosodium salt (500 ppm)
3	3-(N-Formyl-N-hydroxyamino) propylphosphonic acid monosodium salt (500 ppm)
Note)	As test compounds 1*), the corresponding commercial product Gesapax (25% emulsifiable concentrate, Ciba-Geigy) was used.

The test results are as shown below in the table.

Com- pound No.	Plant No.	Hibicidal activity (%)						
		1	2	3	4	5	6	7
1 (Control)		78	82	100	100	100	80	78
2		100	100	100	100	100	100	100
3 (Control)		(22)	(27)	(34)	(40)	(46)	(25)	(28)



The open circle around each figure indicates the occurrence of chlorosis.

Example 2

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The field on which the following weeds were grown, were plotted out in a ratio of 4m<sup>2</sup>/lot.

The following test compound was dissolved in water at the specified application rate and was sprayed onto the whole surface of the weeds at the volume rate of 100 liters water/10ares.

Control lots were treated with water only.

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Test weed code	Test weeds common name	Botanical name	height (cm)
a	Fleabane, Annul	Erigeron annuus L.	60
b	Ohinutade	Polygonum Blumei Meisn	60

5	Test weed code	Test weeds common name	Botanical name	height (cm)
	c	Clover, Red	Trifolium protense L.	40
10	d	Mugwort	Artemisia vulgaris L. var. indica Maxim	40
15	e	Dandelion	Taraxacum Officinale Weber	10-20
20	f	Brome, Rescue	Bromus unioloides Humb. Bonpl. et Kunth	60

30	Test compound No.	Chemical name
35	A	3-(N-Formyl-N-hydroxyamino)- propylphosphonic acid monosodium salt
40	B	2-Methylmercapto-4-ethylamino-6- isopropylamino-s-triazine 1**)
45	C	3-(3,4-Dichlorophenyl)-1,1- dimethylurea 2**)

50 Note) As test compounds 1\*\*) and 2\*\*), the  
corresponding commercial products, Gesapax (25%  
emulsifiable concentrate, Ciba-Geigy) and Karmex  
55 D(78.5% wettable powder, du Pont) were used,  
respectively.

Two weeks after the treatment, herbicidal effect of the test compounds on the weeds were observed and scored on a rating scale of 0 for no effect through - 100 for complete kill.

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Test compound and application rate (g/10a)		Test weed and herbicidal activity (%)					
B	A	a	b	c	d	e	f
0	25	5	5	5	10	10	0
0	50	10	10	10	15	10	10
0	100	20	20	20	30	15	10
62.5	0	0	0	0	0	0	0
125	0	20	20	15	20	10	15
250	0	40	50	60	60	60	55
62.5	25	60	60	50	60	70	60
125	25	70	65	70	80	70	70
250	25	85	80	85	80	85	80
62.5	50	70	70	75	80	70	70
125	50	80	80	90	90	80	80
250	50	90	>90	90	>90	90	90
62.5	100	80	90	80	>90	80	80
125	100	90	>90	>90	>90	90	90
250	100	>90	>90	>90	>90	>90	>90

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5	Test compound and application rate (g/10a)		Test weed and herbicidal activity (%)					
10	C	A	a	b	c	d	e	f
15	0 0 0	25 50 100	5 10 20	5 10 20	5 10 20	10 25 30	10 10 15	0 10 10
20	250 500 750	0 0 0	5 15 40	5 10 30	10 20 40	5 15 30	10 20 40	5 15 30
25	250 500 750	25 25 25	40 60 85	50 60 80	30 40 75	50 50 85	50 55 85	40 60 85
30	250 500 750	50 50 50	60 80 90	70 90 90	50 80 90	70 90 >90	60 90 90	60 85 90
35	250 500 750	100 100 100	85 90 >90	90 >90 >90	80 >90 >90	90 >90 >90	80 >90 >90	70 >90 >90
40								

### 45 Example 3

Seeds of goosefoot were sown in soiled polyethylene pots (20 cm long × 10 cm wide × 10 cm deep) and grown in a glasshouse. Numbers of seedlings (Grown height : 30 cm) were adjusted to 10/pot. Each test compound was dissolved in water at the specified application rate and applied onto the leaves and stocks of Goosefoot. The treated goosefoot were grown in a glass house at 25-30°C for 2 weeks. The weight of the remaining and living portion (above ground) of goose foot was determined and from the obtained data, ratio (%) of the weight of treatment group to that of non-treatment group was calculated. The results are shown in the following table (a).

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(a) Ratio (%) of the remaining weight of the treated plant to that of the non-treated plant

		application rate (g/10a) of the test compound A			
		0	25	50	100
application rate (g/10a) of the test compound B	0	100	95	85	85
	62.5	100	45	25	10
	125	80	20	10	<10
	250	50	10	10	<10

Note: Test compound A: 3-(N-formyl-N-hydroxyamino)-propylphosphonic acid monosodium salt

Test compound B: 2-methylmercapto-4-ethylamino-

6-isopropylamino-s-triazine  
[Gesapax (25% emulsifiable concentrate, Ciba-Geigy)]

The above data were analyzed according to Colby's equation (cf. weeds, Vol, 15, P. 20-22).

Expected value (%) of the remaining weight of the  
plant treated with the combination

Observed value (%) of the  
remaining weight of the  
plant treated with  
compound A only

X

Observed value (%) of  
the remaining weight  
of the plant treated  
with compound B only

=  
100

In case that the expected value (%) is larger than the observed value (%), the combination can be judged to have synergistic effect.

The Colby's expected values (%) obtained by calculating using the above data are shown in the following table (b).

(b) Colby's expected value

		application rate (g/10a) of the test compound A		
		25	50	100
application rate (g/10a) of the test compound B	62.5	95	85	85
	125	76	68	68
	250	47.5	42.5	42.5

As clear from the data listed in the above table (a) and (b), all combinations of the test compound A and B can be judged to have synergistic herbicidal activity.

## Example 4

The following data were obtained in a similar manner to those of Example 3, excepting the test compound C was used in place of the compound B.

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(a) Ratio (%) of the remaining weight of the treated plant to that of the non-treated plant

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		application rate (g/10a) of the test compound A			
		0	25	50	100
application rate (g/10a) of the test compound C	0	100	95	85	85
	250	95	60	40	20
	500	85	45	10	<10
	750	60	20	10	<10

Note: Test compound A is the same compound as defined in Example 3.

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Test compound C : 3-(3,4-dichlorophenyl)-1,1-dimethylurea

[Karmex D (78.5% wettable powder, du Pont)].

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(b) Colby's exypected value (%)

5	application rate (g/10a) of the test compound A						
10	<table><tr><td>25</td><td>50</td><td>100</td></tr></table>				25	50	100
25	50	100					
15	application rate (g/10a) of the test compound C	250	90	81	81		
20		500	81	72	72		
		750	57	51	51		

As clear from the data listed in the above table (a) and (b), all combinations of the test compound A and C can be judged to have synergistic herbicidal activity.

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#### Claims

1. A herbicidal composition comprising fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and diuron, and agronomically acceptable carrier(s).
2. The herbicidal composition of claim 1, which comprises fosmidomycin or salt thereof in combination with ametryn or salt thereof, and agronomically acceptable carrier(s).
3. The herbicidal composition of claim 1, which comprises fosmidomycin or salt thereof in combination with diuron, and agronomically acceptable carrier(s).
4. A method of killing weeds which comprises applying to weed seedlings fosmidomycin or salt thereof in combination with the other herbicide selected from the group of ametryn or salt thereof and diuron.

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